

# Shot noise in non-adiabatically driven nanoscale conductors

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We investigate the noise properties of pump currents through molecular wires and coupled quantum dots. As a model we employ a two level system that is connected to electron reservoirs and is non-adiabatically driven. Concerning the electron-electron interaction, we focus on two limits: non-interacting electrons and strong Coulomb repulsion. While the former case is treated within a Floquet scattering formalism, we derive for the latter case a master equation formalism for the computation of the current and the zero-frequency noise. For a pump operated close to internal resonances, the differences between the non-interacting and the strongly interacting limit turn out to be surprisingly small.

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## 1 Introduction

Recent experiments with coherently coupled quantum dots [1–4] and molecular wires [5, 6] deal with the transport properties of small systems with a discrete level structure. These experimental achievements generated new theoretical interest in the transport properties of nanoscale systems [7, 8]. One particular field of interest is the interplay of transport and electronic excitations by an oscillating gate voltage, a microwave field, or an infrared laser, respectively. Such excitations bear intriguing phenomena like photon-assisted tunnelling [3, 9–16] and the suppression of both the dc current [17, 18] and the zero-frequency noise [19, 20].

A further intriguing phenomenon in this context is electron pumping induced by a cyclic change of the parameters in the absence of any external bias voltage [21–23]. For adiabatically slow driving, the transferred charge per cycle is determined by the area enclosed in parameter space during the cyclic evolution [24, 25]. This implies that the resulting current is proportional to the driving frequency and, thus, suggests that non-adiabatic electron pumping is more effective. For practical applications, it is also desirable that the pump current flows at a sufficiently low noise level. It has been found that adiabatic pumps can be practically noiseless [26]. This happens, however, on the expense of having only a small or even vanishing current [27]. Outside the adiabatic regime, when the driving frequency is close to the internal resonances of the conductor, the current assumes much larger values while its noise nevertheless is clearly sub-Poissonian [28]. Since this prediction of an optimal working point has been made for non-interacting electrons, the question on the influence of Coulomb repulsion arises.

An intuitive description of the electron transport through mesoscopic systems is provided by the Landauer scattering formula [29, 30] and its various generalisations. In this formalism, both the average current [31] and the transport noise characteristics [30, 32] can be expressed in terms of the quantum transmission probabilities of the respective scattering channels. If one heuristically postulates that the current obeys a scattering formula, one should worry whether this complies with the Pauli principle or if it has to be ensured by introducing “blocking factors” [31]. For static conductors the current being the experimentally relevant quantity, is independent of these blocking factors, which renders this question rather academic.

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This is no longer the case when the scattering potential is time-dependent. Then a scattered electron can absorb or emit energy quanta from the driving field, which opens inelastic transport channels [33–35]. So blocking factors indeed can have a net effect on the current, and it has been suggested to test the demand for them experimentally with driven conductors [36, 37]. In order to avoid such conflicts, one should start from a many-particle description. In this spirit, within a Green function approach, a formal solution for the current through a time-dependent conductor has been presented, e.g., in Refs. [36] and [38] without taking advantage of the full Floquet theory for the wire. Nevertheless in some special cases like, e.g., for conductors consisting of only a single level [39, 40] or for the scattering by a piecewise constant potential [41], an explicit solution becomes feasible. A complete Floquet theory provides in addition to a current formula a prescription for the computation of the Green function [16, 42].

The spectral density of the current fluctuations has been derived for the low-frequency ac conductance [43, 44] and the scattering by a slowly time-dependent potential [45]. For arbitrary driving frequencies, the noise has been characterised by its zero-frequency component [19]. A remarkable feature of the current noise in the presence of time-dependent fields is its dependence on the phase of the transmission amplitudes [19, 45]. By clear contrast, both the current in the driven case [19] and the noise in the static case [30] depend solely on transmission probabilities.

When electron-electron interactions beyond the mean-field level become relevant, the direct application of a Landauer-like theory is no longer possible and one has to resort to other methods like, e.g., a master equation description for the reduced density operator of the wire [46–49]. For time-dependent conductors, this enables a rather efficient treatment of transport problems after decomposing the wire density operator into a Floquet basis. Then it is possible to study relatively large driven conductors [16] and to include also electron-electron interactions [50, 51] and electron-phonon interactions [52]. For the computation of the current fluctuations, one can employ a generalised master equation that resolves the number of the transported electrons. This degree of freedom is traced out after introducing a counting variable [53]. For various static transport problems, this approach has been followed by several groups [54–60].

After introducing our model, we review in Sec. 2 the Floquet scattering theory for the computation of the current and the zero-frequency noise. In Sec. 3, we derive a master equation approach which is applicable also in the presence of electron-electron interactions. These formalisms are in Sec. 4 employed for investigating the influence of Coulomb repulsion on the noise in non-adiabatic electron pumps.

### 1.1 Wire-lead model

A frequently used model for a nanoscale conductors like molecular wires or coupled quantum dots is sketched in Fig. 1. It is described by the time-dependent Hamiltonian

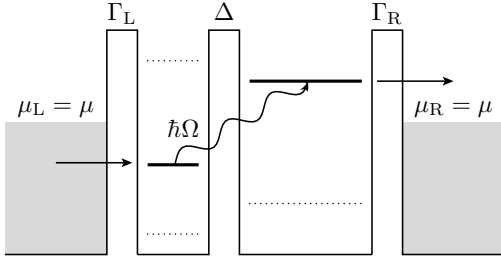
$$H(t) = H_{\text{wire}}(t) + H_{\text{leads}} + H_{\text{contacts}}, \quad (1)$$

where the different terms correspond to the central conductor (“wire”), electron reservoirs (“leads”), and the wire-lead couplings, respectively. We focus on the regime of coherent quantum transport where the main physics at work occurs on the wire itself. In doing so, we neglect other possible influences originating from driving-induced hot electrons in the leads and dissipation on the wire. Then, the wire Hamiltonian reads in a tight-binding approximation with  $N$  orbitals  $|n\rangle$

$$H_{\text{wire}}(t) = \sum_{n,n',s,s'} H_{nn'}(t) c_{ns}^\dagger c_{n's'} + H_{\text{interaction}}. \quad (2)$$

For a molecular wire, this constitutes the so-called Hückel description where each site corresponds to one atom. The fermion operators  $c_{ns}$ ,  $c_{ns}^\dagger$  annihilate and create, respectively, an electron with spin  $s = \uparrow, \downarrow$  in the orbital  $|n\rangle$ . The influence of an applied ac field or an oscillating gate voltage with frequency  $\Omega = 2\pi/\mathcal{T}$  results in a periodic time-dependence of the wire Hamiltonian:  $H_{nn'}(t+\mathcal{T}) = H_{nn'}(t)$ . For the interaction Hamiltonian, we assume a capacitor model, so that

$$H_{\text{interaction}} = \frac{U}{2} N_{\text{wire}} (N_{\text{wire}} - 1) \quad (3)$$



**Fig. 1** Level structure of a double quantum dot with  $N = 2$  orbitals. The terminating sites are coupled to leads with chemical potential  $\mu_L$  and  $\mu_R = \mu_L + eV$ , respectively.

where  $N_{\text{wire}} = \sum_{ns} c_{ns}^\dagger c_{ns}$  describes the number of electrons on the wire. Below we shall focus on two limits, namely the interaction-free case  $U = 0$  and strong interaction,  $U \rightarrow \infty$ , which finally means that the Coulomb repulsion is so strong that only states with zero or one excess electron play a role.

The leads are modelled by ideal electron gases,

$$H_{\text{leads}} = \sum_{q,s} \epsilon_q (c_{Lqs}^\dagger c_{Lqs} + c_{Rqs}^\dagger c_{Rqs}), \quad (4)$$

where  $c_{Lq}^\dagger$  ( $c_{Rq}^\dagger$ ) creates an electron in the state  $|Lq\rangle$  ( $|Rq\rangle$ ) in the left (right) lead. The wire-lead tunnelling Hamiltonian

$$H_{\text{contacts}} = \sum_{q,s} \left( V_{Lqs} c_{Lqs}^\dagger c_{1s} + V_{Rqs} c_{Rqs}^\dagger c_{Ns} \right) + \text{h.c.} \quad (5)$$

establishes the contact between the sites  $|1\rangle$ ,  $|N\rangle$  and the respective lead. This tunnelling coupling is described by the spectral density

$$\Gamma_\ell(\epsilon) = 2\pi \sum_q |V_{\ell q}|^2 \delta(\epsilon - \epsilon_q) \quad (6)$$

of lead  $\ell = L, R$ . In the following, we restrict ourselves to the so-called wide-band limit in which the spectral density is assumed to be energy-independent,  $\Gamma_\ell(\epsilon) \rightarrow \Gamma_\ell$ .

To fully specify the dynamics, we choose as an initial condition for the left/right lead a grand-canonical electron ensemble at temperature  $T$  and electro-chemical potential  $\mu_{L/R}$ . Thus, the initial density matrix reads

$$\rho_0 \propto e^{-(H_{\text{leads}} - \mu_L N_L - \mu_R N_R)/k_B T}, \quad (7)$$

where  $N_\ell = \sum_{qs} c_{\ell qs}^\dagger c_{\ell qs}$  is the number of electrons in lead  $\ell$  and  $k_B T$  denotes the Boltzmann constant times temperature. An applied voltage  $V$  maps to a chemical potential difference  $\mu_R - \mu_L = eV$  with  $-e$  being the electron charge. Then, at initial time  $t_0$ , the only nontrivial expectation values of the wire operators read  $\langle c_{\ell' q' s'}^\dagger c_{\ell q s} \rangle = f_\ell(\epsilon_q) \delta_{\ell' \ell} \delta_{q' q} \delta_{s' s}$  where  $f_\ell(\epsilon) = (1 + \exp[(\epsilon - \mu_\ell)/k_B T])^{-1}$  denotes the Fermi function.

## 1.2 Charge, current, and current fluctuations

To avoid the explicit appearance of commutators in the definition of correlation functions, we perform the derivation of the central transport quantities in the Heisenberg picture. As a starting point we choose the operator

$$Q_\ell(t) = eN_\ell(t) - eN_\ell(t_0) \quad (8)$$

which describes the charge accumulated in lead  $\ell$  with respect to the initial state. Due to total charge conservation,  $Q_\ell$  equals the net charge transmitted across the contact  $\ell$ ; its time derivative defines the

corresponding current

$$I_\ell(t) = \frac{d}{dt} Q_\ell(t). \quad (9)$$

The current noise is described by the symmetrised correlation function

$$S_\ell(t, t') = \frac{1}{2} \langle [\Delta I_\ell(t), \Delta I_\ell(t')]_+ \rangle \quad (10)$$

of the current fluctuation operator  $\Delta I_\ell(t) = I_\ell(t) - \langle I_\ell(t) \rangle$ , where the anticommutator  $[A, B]_+ = AB + BA$  ensures hermiticity. At long times,  $S_\ell(t, t') = S_\ell(t + T, t' + T)$  shares the time-periodicity of the driving [42]. Therefore, it is possible to characterise the noise level by the zero-frequency component of  $S_\ell(t, t - \tau)$  averaged over the driving period,

$$\bar{S}_\ell = \frac{1}{T} \int_0^T dt \int_{-\infty}^{\infty} d\tau S_\ell(t, t - \tau). \quad (11)$$

Moreover for two-terminal devices,  $\bar{S}_\ell$  is independent of the contact  $\ell$ , i.e.,  $\bar{S}_L = \bar{S}_R \equiv \bar{S}$ .

The evaluation of the zero-frequency noise  $\bar{S}$  directly from its definition (11) can be tedious due to the explicit appearance of both times,  $t$  and  $t - \tau$ . This inconvenience can be circumvented by employing the relation

$$\frac{d}{dt} \left( \langle Q_\ell^2(t) \rangle - \langle Q_\ell(t) \rangle^2 \right) = 2 \int_0^{\infty} d\tau S_\ell(t, t - \tau), \quad (12)$$

which follows from the integral representation of Eqs. (8) and (9),  $Q_\ell(t) = \int_{t_0}^t dt' I_\ell(t')$ , in the limit  $t_0 \rightarrow -\infty$ . By averaging Eq. (12) over the driving period and using  $S(t, t - \tau) = S(t - \tau, t)$ , we obtain

$$\bar{S} = \left\langle \frac{d}{dt} \langle \Delta Q_\ell^2(t) \rangle \right\rangle_t, \quad (13)$$

where  $\Delta Q_\ell = Q_\ell - \langle Q_\ell \rangle$  denotes the charge fluctuation operator and  $\langle \dots \rangle_t$  the time average. The fact that the time average can be evaluated from the limit  $\bar{S} = \lim_{t_0 \rightarrow -\infty} \langle \Delta Q_\ell^2(t) \rangle / (t - t_0) > 0$  allows to interpret the zero-frequency noise as the “charge diffusion coefficient”. As a dimensionless measure for the *relative* noise strength, we employ the so-called Fano factor [61]

$$F = \frac{\bar{S}}{e|\bar{I}|}, \quad (14)$$

which can provide information about the nature of the transport mechanism [30, 62]. Here,  $\bar{I}$  denotes the time-average of the current expectation value  $\langle I_\ell(t) \rangle$ . Historically, the zero-frequency noise (11) contains a factor 2, i.e.  $\bar{S}' = 2\bar{S}$ , resulting from a different definition of the Fourier transform. Then, the Fano factor is defined as  $F = \bar{S}' / 2e|\bar{I}|$ .

### 1.3 Full counting statistics

A more complete picture of the current fluctuations beyond second order correlations is provided by the full counting statistics. It is determined by the moment generating function

$$\phi(\chi, t) = \langle e^{i\chi N_L} \rangle_t \quad (15)$$

and allows the direct computation of the  $k$ th moment of the charge in the left lead via the relation

$$\langle Q_L^k(t) \rangle = e^k \frac{\partial^k \phi(\chi)}{\partial (i\chi)^k} \phi(\chi, t) \Big|_{\chi=0}. \quad (16)$$

Subtracting from the moments the trivial contributions that depend on a shift of the initial values, one obtains the cumulants. They are defined and generated via the so-called cumulant generating function  $\ln \phi(\chi, t)$  which replaces  $\phi$  in Eq. (16) [63], so that the  $k$ th cumulant reads

$$C_k = e^k \frac{\partial^k \phi(\chi)}{\partial (i\chi)^k} \ln \phi(\chi, t) \Big|_{\chi=0}. \quad (17)$$

In a continuum limit for the leads, both the moments and the cumulants diverge as a function of time, and one focusses on the rates at which these quantities change in the long-time limit. This establishes between the first two cumulants and  $I(t)$  and  $S(t)$  the relations

$$I(t) = -ie \frac{\partial}{\partial \chi} \dot{C}(\chi, t) \Big|_{\chi=0}, \quad (18)$$

$$S(t) = -e^2 \frac{\partial^2}{\partial \chi^2} \dot{C}(\chi, t) \Big|_{\chi=0}. \quad (19)$$

For driven systems, these quantities are time-dependent even in the asymptotic limit and, thus, we characterise the transport by the corresponding averages over one driving period. Then expressions (18) and (19) become identical to the previously defined time averages  $\bar{I}$  and  $\bar{S}$ , respectively. Herein we restrict ourselves to the computation of the first and the second cumulant, despite the fact that also higher-order cumulants can be measured [64, 65].

## 2 Floquet scattering theory

We now derive from the model described in Section 1.1 in the absence of electron-electron interactions expressions for both the current through the wire and the associated noise by solving the corresponding Heisenberg equations of motions. Since for  $U = 0$ , the both spin directions contribute independently to the current, we ignore the spin index which means that we consider the current per spin projection. We start from the equations of motion for the annihilation operators in lead  $\ell$ ,

$$\dot{c}_{\ell q} = -\frac{i}{\hbar} \epsilon_{\ell q} c_{\ell q} - \frac{i}{\hbar} V_{\ell q} c_{n_\ell}, \quad (20)$$

which are straightforwardly integrated to read

$$c_{\ell q}(t) = c_{\ell q}(t_0) e^{-i\epsilon_{\ell q}(t-t_0)/\hbar} - \frac{i}{\hbar} V_{\ell q} \int_0^{t-t_0} d\tau e^{-i\epsilon_{\ell q}\tau/\hbar} c_{n_\ell}(t-\tau), \quad (21)$$

where  $n_\ell$  denotes the molecular wire site attached to lead  $\ell$ , i.e.,  $n_L = 1$  and  $n_R = N$ . Inserting (21) into the Heisenberg equations for the wire operators yields in the asymptotic limit  $t_0 \rightarrow -\infty$

$$\dot{c}_{n_\ell}(t) = -\frac{i}{\hbar} \sum_{n'} H_{n_\ell, n'}(t) c_{n'}(t) - \frac{1}{2\hbar} \Gamma_\ell c_{n_\ell}(t) + \xi_\ell(t), \quad (22)$$

$$\dot{c}_n(t) = -\frac{i}{\hbar} \sum_{n'} H_{nn'}(t) c_{n'}(t), \quad n = 2, \dots, N-1. \quad (23)$$

For a energy-dependent spectral density  $\Gamma_\ell = \Gamma_\ell(\epsilon)$ , the dissipative part of the Heisenberg equation (22) acquires a memory kernel which complicates not only its solution but also the derivation of a current formula. For details, we refer the reader to Ref. [16].

The influence of the operator-valued Gaussian noise

$$\xi_\ell(t) = -\frac{i}{\hbar} \sum_q V_{\ell q}^* e^{-i\epsilon_{\ell q}(t-t_0)/\hbar} c_{\ell q}(t_0) \quad (24)$$

is fully specified by the expectation values  $\langle \xi_\ell(t) \rangle = 0$  and

$$\langle \xi_{\ell'}^\dagger(t') \xi_\ell(t) \rangle = \delta_{\ell\ell'} \frac{\Gamma_\ell}{2\pi\hbar^2} \int d\epsilon e^{-i\epsilon(t-t')/\hbar} f_\ell(\epsilon), \quad (25)$$

which follow directly from the definition (24) and the initial conditions (7). It is convenient to define the Fourier representation of the noise operator,  $\xi_\ell(\epsilon) = \int dt \exp(i\epsilon t/\hbar) \xi_\ell(t)$  whose correlation function

$$\langle \xi_\ell^\dagger(\epsilon) \xi_{\ell'}(\epsilon') \rangle = 2\pi\Gamma_\ell f_\ell(\epsilon) \delta(\epsilon - \epsilon') \delta_{\ell\ell'} \quad (26)$$

follows directly from Eq. (25).

### 2.1 Retarded Green function

The equations of motion (22) and (23) represent a set of linear inhomogeneous equations and, thus, can be solved with the help of a retarded Green function  $G(t, t') = -(i/\hbar) U(t, t') \theta(t - t')$ , which obeys

$$\left( i\hbar \frac{d}{dt} - \mathcal{H}(t) + \frac{i}{2}\Gamma \right) G(t, t') = \delta(t - t'), \quad (27)$$

where  $\Gamma(t) = |1\rangle\Gamma_L(t)\langle 1| + |N\rangle\Gamma_R(t)\langle N|$  and  $\mathcal{H}(t)$  is the one-particle Hamiltonian corresponding to Eq. (2). At this stage, it is important to note that the propagator of the homogeneous equations obeys  $U(t, t') = U(t + T, t' + T)$ . Accordingly, the Fourier representation of the retarded Green function

$$G(t, \epsilon) = -\frac{i}{\hbar} \int_0^\infty d\tau e^{i\epsilon\tau/\hbar} U(t, t - \tau) = G(t + T, \epsilon) = \sum_{k=-\infty}^{\infty} e^{-ik\Omega t} G^{(k)}(\epsilon) \quad (28)$$

is also  $T$ -periodic in the time argument, so that it can be represented as a Fourier series. Physically, the Fourier coefficients  $G^{(k)}(\epsilon)$  describe the propagation of an electron with initial energy  $\epsilon$  under the absorption (emission) of  $|k|$  photons for  $k > 0$  ( $k < 0$ ). In the limiting case of a time-independent situation, all sideband contributions with  $k \neq 0$  vanish and  $G(t, \epsilon)$  becomes time-independent and identical to  $G^{(0)}(\epsilon)$ . From the definition (27) of the Green function and its Fourier representation (28), it can be shown that the solution of the Heisenberg equations (22), (23) reads

$$c_n(t) = \frac{i}{2\pi} \sum_\ell \int d\epsilon e^{-i\epsilon t/\hbar} G_{n,n_\ell}(t, \epsilon) \xi_\ell(\epsilon), \quad (29)$$

where we have defined  $G_{n,n_\ell}(t, \epsilon) = \langle n | G(t, \epsilon) | n_\ell \rangle$ .

Below, we need for the elimination of back-scattering terms the relation

$$G^\dagger(t, \epsilon') - G(t, \epsilon) = \left( i\hbar \frac{d}{dt} - \epsilon' + \epsilon + i\Gamma \right) G^\dagger(t, \epsilon') G(t, \epsilon). \quad (30)$$

A proof starts from the definition of the Green function, Eq. (27). By Fourier transformation with respect to  $t'$ , we obtain the relation

$$\left( i\hbar \frac{d}{dt} + \epsilon - \mathcal{H}(t) + \frac{i}{2}\Gamma \right) G(t, \epsilon) = \mathbf{1}, \quad (31)$$

which we multiply by  $G^\dagger(t, \epsilon)$  from the left. The difference between the resulting expression and its hermitian adjoint with  $\epsilon$  and  $\epsilon'$  interchanged is relation (30).

## 2.2 Current through the driven molecular wire

Owing to charge conservation, the (net) current flowing from lead  $\ell$  into the molecular wire is determined by the negative time derivative of the charge in lead  $\ell$ . Thus, the current operator reads  $I_\ell = ie[H(t), N_\ell]/\hbar$ , where  $N_\ell = \sum_q c_{\ell q}^\dagger c_{\ell q}$  denotes the corresponding electron number and  $-e$  the electron charge. From Eqs. (21) and (24) then follows

$$I_L(t) = \frac{e\Gamma_L}{\hbar} \int_0^\infty d\tau \{c_1^\dagger(t)c_1(t-\tau) + c_1^\dagger(t-\tau)c_1(t)\} - e\{c_1^\dagger(t)\xi_L(t) + \xi_L^\dagger(t)c_1(t)\}. \quad (32)$$

This operator-valued expression for the time-dependent current is a convenient starting point for the evaluation of expectation values like the dc and ac current and the current noise.

### 2.2.1 Time-average current

To obtain the current  $\langle I_L(t) \rangle$ , we insert the solution (29) of the Heisenberg equation into the current operator (32) and use the expectation values (26). The resulting expression

$$\langle I_L(t) \rangle = \frac{e\Gamma_L}{\hbar} \sum_\ell \Gamma_\ell \text{Im} \int d\epsilon f_\ell(\epsilon) e^{i\epsilon\tau/\hbar} G_{1\ell}^*(t, \epsilon) G_{1\ell}(t-\tau, \epsilon) + 2e\Gamma_L \text{Im} \int d\epsilon f_L(\epsilon) G_{11}(t, \epsilon) \quad (33)$$

still contains back-scattering terms  $G_{11}$  and, thus, is not of a “scattering form”. Indeed, bringing (33) into a form that resembles the static current formula requires some tedious algebra. Such a derivation has been presented for the linear conductance of time-independent systems [66], for tunnelling barriers [67] and mesoscopic conductors [68] in the static case for finite voltage, and for a wire consisting of levels that couple equally strong to both leads [38]. For the periodically time-dependent case in the absence of electron-electron interactions, such an expression has been *derived* only recently [19,42].

Inserting the matrix element  $\langle 1 | \dots | 1 \rangle$  of equation (30) eliminates the back-scattering terms and yields for the time-dependent current the expression

$$\langle I_L(t) \rangle = \frac{e}{\hbar} \int d\epsilon \{T_{LR}(t, \epsilon) f_R(\epsilon) - T_{RL}(t, \epsilon) f_L(\epsilon)\} - \frac{d}{dt} q_L(t) \quad (34)$$

where  $q_L(t)$  denotes the charge oscillating between the left lead and the wire. Obviously, since  $q_L(t)$  is time-periodic and bounded, its time derivative cannot contribute to the average current. The time-dependent current is determined by the time-dependent transmission

$$T_{LR}(t, \epsilon) = \Gamma_L \Gamma_R \text{Re} \int_0^\infty d\tau e^{i\epsilon\tau/\hbar} G_{1N}^*(t, \epsilon) G_{1N}(t-\tau, \epsilon). \quad (35)$$

The corresponding expression for  $T_{RL}(t, \epsilon)$  follows from the replacement  $(L, 1) \leftrightarrow (R, N)$ . We emphasise that expression (34) obeys the form of the current formula for a static conductor within a scattering formalism. In particular, consistent with Refs. [31,36], no “Pauli blocking factors”  $(1 - f_\ell)$  appear in our derivation.

The dc current obtained from (34) by time-averaging can be written in an even more compact form if we insert for the Green function the Fourier representation (28). This results in

$$\bar{I} = \frac{e}{\hbar} \sum_k \int d\epsilon \{T_{LR}^{(k)}(\epsilon) f_R(\epsilon) - T_{RL}^{(k)}(\epsilon) f_L(\epsilon)\}, \quad (36)$$

where

$$T_{LR}^{(k)}(\epsilon) = \Gamma_L \Gamma_R |G_{1N}^{(k)}(\epsilon)|^2, \quad (37)$$

$$T_{RL}^{(k)}(\epsilon) = \Gamma_R \Gamma_L |G_{N1}^{(k)}(\epsilon)|^2, \quad (38)$$

denote the transmission probabilities for electrons from the right to the left lead and vice versa, respectively, with initial energy  $\epsilon$  and final energy  $\epsilon + k\hbar\Omega$ , i.e., the probability for an scattering event under the absorption (emission) of  $|k|$  photons if  $k > 0$  ( $k < 0$ ).

For a static situation, all contributions with  $k \neq 0$  vanish and  $T_{LR}^{(0)}(\epsilon) = T_{RL}^{(0)}(\epsilon)$ . Therefore, it is possible to write the current (36) as a product of a single transmission  $T(\epsilon)$ , which is independent of the direction, and the difference of the Fermi functions,  $f_R(\epsilon) - f_L(\epsilon)$ . We emphasise that in the driven case this no longer holds true.

### 2.2.2 Noise power

In order to derive a related expression for the time-averaged current-current correlation function (11), we insert the current operator (32) and the solution (29) of the Heisenberg equations of motion. Then, we again employ relation (30) and the shorthand notation  $\epsilon_k = \epsilon + k\hbar\Omega$ , so that we finally obtain

$$\begin{aligned} \bar{S} = & \frac{e^2}{h} \sum_k \int d\epsilon \left\{ \Gamma_R \Gamma_R \left| \sum_{k'} \Gamma_L(\epsilon_{k'}) G_{1N}^{(k'-k)}(\epsilon_k) [G_{1N}^{(k')}(\epsilon)]^* \right|^2 f_R(\epsilon) \bar{f}_R(\epsilon_k) \right. \\ & + \Gamma_R \Gamma_L \left| \sum_{k'} \Gamma_L G_{1N}^{(k'-k)}(\epsilon_k) [G_{11}^{(k')}(\epsilon)]^* - i G_{1N}^{(-k)}(\epsilon_k) \right|^2 f_L(\epsilon) \bar{f}_R(\epsilon_k) \Big\} \\ & + \text{same terms with the replacement } (L, 1) \leftrightarrow (R, N). \end{aligned} \quad (39)$$

### 2.2.3 Floquet decomposition in the wide-band limit

Solving the equations of motion (27) for the Green function is equivalent to computing a complete set of solutions for the equation

$$i\hbar \frac{d}{dt} |\psi(t)\rangle = \left( \mathcal{H}_{\text{wire}}(t) - \frac{i}{2} \Gamma \right) |\psi(t)\rangle, \quad (40)$$

which is linear and possesses time-dependent,  $\mathcal{T}$ -periodic coefficients. Thus, it is possible to construct a complete set of solutions with the Floquet ansatz

$$|\psi_\alpha(t)\rangle = \exp[(-i\epsilon_\alpha/\hbar - \gamma_\alpha)t] |u_\alpha(t)\rangle, \quad (41)$$

$$|u_\alpha(t)\rangle = \sum_k \exp(-ik\Omega t) |u_{\alpha,k}\rangle. \quad (42)$$

The so-called Floquet states  $|u_\alpha(t)\rangle$  obey the time-periodicity of  $\mathcal{H}_{\text{wire}}(t)$  and have been decomposed into a Fourier series. In a Hilbert space that is extended by a periodic time coordinate, the so-called Sambe space [69], they obey the Floquet eigenvalue equation [70, 71]

$$\left( \mathcal{H}_{\text{wire}}(t) - i\Sigma - i\hbar \frac{d}{dt} \right) |u_\alpha(t)\rangle = (\epsilon_\alpha - i\hbar\gamma_\alpha) |u_\alpha(t)\rangle. \quad (43)$$

Due to the Brillouin zone structure of the Floquet spectrum [69, 70, 72], it is sufficient to compute all eigenvalues of the first Brillouin zone,  $-\hbar\Omega/2 < \epsilon_\alpha \leq \hbar\Omega/2$ . Since the operator on the l.h.s. of Eq. (43) is non-Hermitian, the eigenvalues  $\epsilon_\alpha - i\hbar\gamma_\alpha$  are generally complex-valued and the (right) eigenvectors are not mutually orthogonal. Thus, to determine the propagator, we need to solve also the adjoint Floquet equation yielding again the same eigenvalues but providing the adjoint eigenvectors  $|u_\alpha^+(t)\rangle$ . It can be shown that the Floquet states  $|u_\alpha(t)\rangle$  together with the adjoint states  $|u_\alpha^+(t)\rangle$  form at equal times a complete bi-orthogonal basis:  $\langle u_\alpha^+(t) | u_\beta(t) \rangle = \delta_{\alpha\beta}$  and  $\sum_\alpha |u_\alpha(t)\rangle \langle u_\alpha^+(t)| = \mathbf{1}$ . A proof requires to account for the time-periodicity of the Floquet states since the eigenvalue equation (43) holds in a Hilbert space extended by a periodic time coordinate [70, 73].



Using the Floquet equation (43), it is straightforward to show that the propagator can be written as

$$U(t, t') = \sum_{\alpha} e^{-i(\epsilon_{\alpha}/\hbar - i\gamma_{\alpha})(t-t')} |u_{\alpha}(t)\rangle \langle u_{\alpha}^+(t')|, \quad (44)$$

where the sum runs over all Floquet states within one Brillouin zone. Consequently, the Fourier coefficients of the Green function read

$$G^{(k)}(\epsilon) = -\frac{i}{\hbar} \int_0^T \frac{dt}{T} e^{ik\Omega t} \int_0^{\infty} d\tau e^{i\epsilon\tau/\hbar} U(t, t-\tau) \quad (45)$$

$$= \sum_{\alpha, k'} \frac{|u_{\alpha, k'+k}\rangle \langle u_{\alpha, k'}^+|}{\epsilon - (\epsilon_{\alpha} + k'\hbar\Omega - i\hbar\gamma_{\alpha})}. \quad (46)$$

In general, the Floquet equation (43) has to be solved numerically. In the zero temperature limit considered here, the Fermi functions in the expressions for the average current (36) and the zero-frequency noise (39) become step functions. Therefore, the integrands are rational functions and the remaining energy integrals can be performed analytically.

### 3 Master equation approach

In the presence of electron-electron interactions, an exact treatment of the electron transport within a scattering theory is no longer possible and a master equation formalism can be an appropriate tool for the computation of currents [13, 16, 46, 51, 52]. Recently, master equations have been established for the computation of current noise of various static conductors as well [53–60]. In the following, we develop such an approach for the case of periodically time-dependent conductors.

#### 3.1 Perturbation theory and reduced density operator

We start our derivation of a master equation formalism from the Liouville-von Neumann equation  $i\hbar\dot{\mathcal{R}}(t) = [H(t), \mathcal{R}(t)]$  for the total density operator  $\mathcal{R}(t)$ . By standard techniques we obtain the exact equation of motion

$$\frac{d}{dt}\tilde{\mathcal{R}}(t) = -\frac{i}{\hbar}[\tilde{H}_{\text{wire-leads}}(t), \mathcal{R}(0)] - \frac{1}{\hbar^2} \int_0^{\infty} d\tau [H_{\text{wire-leads}}, [\tilde{H}_{\text{wire-leads}}(t-\tau, t), \mathcal{R}(t)]], \quad (47)$$

where the tilde denotes the interaction picture with respect to the lead and the wire Hamiltonian,  $\tilde{X}(t, t') = U_0^\dagger(t, t') X U_0(t, t')$ , and  $U_0$  is the propagator without the coupling. Below we will employ Floquet theory in order to obtain explicit expressions for these operators.

As already discussed above, the moment generating function  $\phi(\chi) = \langle \exp(i\chi N_L) \rangle$  contains the full information about the counting statistics. For its explicit computation, we define in the Hilbert space of the wire the operator

$$\mathcal{F}(\chi, t) = \text{tr}_{\text{leads}} \{ e^{i\chi N_L} \mathcal{R}(t) \}, \quad (48)$$

whose limit  $\chi \rightarrow 0$  obviously is the reduced density operator of the wire,  $\mathcal{F}(0, t) = \rho$ . After tracing out the wire degrees of freedom,  $\mathcal{F}$  becomes the moment generating function  $\phi(\chi) = \text{tr}_{\text{wire}} \mathcal{F}$ . It will prove convenient to decompose  $\mathcal{F}$  into a Taylor series,

$$\mathcal{F} = \rho + \sum_{k=1}^{\infty} \frac{(i\chi)^k}{k!} \mathcal{F}_k, \quad (49)$$

where the coefficients  $\mathcal{F}_k = \text{tr}_{\text{leads}} (N_L^k \mathcal{R})$  provide direct access to the moments  $\langle N_L^k \rangle = \text{tr}_{\text{wire}} \mathcal{F}_k$ .

Our strategy is now to derive from the master equation (47) for the full density operator an equation of motion for the  $\mathcal{F}_k$ . For that purpose, we transform the master equation for  $\tilde{\mathcal{R}}$  back to the Schrödinger picture and multiply it from the left by the operator  $\exp(i\chi N_L)$ . By tracing out the lead degrees of freedom and using the commutation relations  $[N_L, V] = V$  and  $[N_L, V^\dagger] = -V^\dagger$ , we obtain

$$\frac{d}{dt}\mathcal{F}(\chi, t) = \{\mathcal{L} + (e^{i\chi} - 1)\mathcal{J}_+ + (e^{-i\chi} - 1)\mathcal{J}_-\}\mathcal{F}(\chi, t). \quad (50)$$

In order to achieve this compact notation, we have defined the superoperators  $\mathcal{J}_\pm$  and the time-dependent Liouville operator

$$\begin{aligned} \mathcal{L}(t) X = & -\frac{i}{\hbar}[H_{\text{wire}}(t), X] \\ & + \frac{\Gamma_L}{2\pi} \int_0^\infty d\tau \int d\epsilon \left[ e^{i\epsilon\tau} (-c_1 \tilde{c}_1^\dagger X f_L(\epsilon) + \tilde{c}_1^\dagger X c_1 f_L(\epsilon) - X \tilde{c}_1^\dagger c_1 \bar{f}_L(\epsilon) + c_1 X \tilde{c}_1^\dagger \bar{f}_L(\epsilon)) \right. \\ & \quad \left. + e^{-i\epsilon\tau} (-X \tilde{c}_1 c_1^\dagger f_L(\epsilon) + c_1^\dagger X \tilde{c}_1 f_L(\epsilon) - c_1^\dagger \tilde{c}_1 X \bar{f}_L(\epsilon) + \tilde{c}_1 X c_1^\dagger \bar{f}_L(\epsilon)) \right] \\ & + \text{same terms with the replacement } 1, L \rightarrow N, R, \end{aligned} \quad (51)$$

which also determines the time-evolution of the reduced density operator,  $\dot{\rho} = \mathcal{L}(t)\rho$ . The tilde denotes the interaction picture operator  $\tilde{c} = \tilde{c}(t, t - \tau)$  and  $f_\ell$  the Fermi function of lead  $\ell$ , while  $\bar{f}_\ell = 1 - f_\ell$ . The current operators

$$\mathcal{J}_+(t) X = \frac{\Gamma_L}{2\pi} \int_0^\infty d\tau \int d\epsilon (e^{i\epsilon\tau} \tilde{c}_1^\dagger X c_1 + e^{-i\epsilon\tau} c_1^\dagger X \tilde{c}_1) f_L(\epsilon), \quad (52)$$

$$\mathcal{J}_-(t) X = \frac{\Gamma_L}{2\pi} \int_0^\infty d\tau \int d\epsilon (e^{i\epsilon\tau} c_1 X \tilde{c}_1^\dagger + e^{-i\epsilon\tau} \tilde{c}_1 X c_1^\dagger) \bar{f}_L(\epsilon), \quad (53)$$

describe the tunnelling of an electron from the left lead to the wire and the opposite process, respectively. Note that these superoperators still contain a non-trivial time-dependence stemming from the interaction-picture representation of the creation and annihilation operators of wire electrons.

### 3.2 Computation of moments and cumulants

For computation of the current (18) and the zero-frequency noise (19), we generalise the approach of Ref. [56] to the time-dependent case. Since we restrict the noise characterisation to the Fano factor, it is sufficient to compute the long-time behaviour of the first and the second moment of the electron number in the left lead. This information is fully contained in the time-derivative of the operator  $\mathcal{F}$  up to second order in  $\chi$ , for which we obtain by Taylor expansion of the equation of motion (50) the hierarchy

$$\dot{\rho} = \mathcal{L}(t)\rho, \quad (54)$$

$$\dot{\mathcal{F}}_1 = \mathcal{L}(t)\mathcal{F}_1 + (\mathcal{J}_+(t) + \mathcal{J}_-(t))\rho, \quad (55)$$

$$\dot{\mathcal{F}}_2 = \mathcal{L}(t)\mathcal{F}_2 + 2(\mathcal{J}_+(t) + \mathcal{J}_-(t))\mathcal{F}_1 + (\mathcal{J}_+(t) - \mathcal{J}_-(t))\rho. \quad (56)$$

The first equation determines the time-evolution of the reduced density operator, which in the long-time limit becomes the stationary solution  $\rho_0(t)$ . Note that for a driven system, it generally is time-dependent. Replacing in Eq. (55)  $\rho$  by  $\rho_0$  and using the fact that  $\text{tr}_{\text{wire}} \mathcal{L}X = 0$  for any operator  $X$ , we obtain the stationary current

$$I(t) = -e \text{tr}_{\text{wire}} \dot{\mathcal{F}}_1 = -e \text{tr}_{\text{wire}} (\mathcal{J}_+ + \mathcal{J}_-)\rho_0(t). \quad (57)$$

The dc current follows simply by averaging over one driving period and one ends up with the current formula of Ref. [74].

The computation of  $\mathcal{F}_1(t)$  is hindered by the fact that the inverse of a Liouvillian generally does not exist. For static systems this is obvious from the fact that the stationary solution fulfils  $\mathcal{L}\rho_0 = 0$ , which implies that  $\mathcal{L}$  is singular. This unfortunately also complicates the computation of the second cumulant and we proceed in the following way: We start from Eq. (12) which relates the zero frequency noise to the charge fluctuation in the leads and write the time derivative of the first and the second moment of the electron number in the left lead by the operators  $\dot{\mathcal{F}}_{1,2}$ . From the equations of motion (55) and (56), we then find  $S = e^2 \text{tr}_{\text{wire}} \{2(\mathcal{J}_+ + \mathcal{J}_- - I)\mathcal{F}_1 - (\mathcal{J}_+ - \mathcal{J}_-)\rho\}$ , where we again used the relation  $\text{tr}_{\text{wire}} \mathcal{L}X = 0$ . An important observation is now that the first part of this expression vanishes for  $\mathcal{F}_1 \propto \rho_0$ , which can easily be demonstrated by inserting the current expectation value (57). Since  $\rho_0 \text{tr}_{\text{wire}}$  acts as a projector onto the stationary solution  $\rho_0$ , we can define the “perpendicular” part

$$\mathcal{F}_{1\perp} = \mathcal{F}_1 - \rho_0 \text{tr}_{\text{wire}} \mathcal{F}_1, \quad (58)$$

which fulfils the relation  $\text{tr}_{\text{wire}} \mathcal{F}_{1\perp} = 0$  and obeys the equation of motion

$$\dot{\mathcal{F}}_{1\perp} = \mathcal{L}(t) \mathcal{F}_{1\perp} + (\mathcal{J}_+(t) + \mathcal{J}_-(t) - I(t))\rho_0(t), \quad (59)$$

We will see below that in contrast to  $\mathcal{F}_1$ , the long-time limit of the traceless  $\mathcal{F}_{1\perp}$  can be computed directly from the equation of motion (59). Upon inserting Eq. (58) into the equation of motion (56), we finally obtain for the still time-dependent “charge diffusion coefficient” the expression

$$S(t) = e^2 \text{tr}_{\text{wire}} \{2(\mathcal{J}_+ + \mathcal{J}_-)\mathcal{F}_{1\perp} - (\mathcal{J}_+ - \mathcal{J}_-)\rho_0\}, \quad (60)$$

whose time-average finally provides the Fano factor  $F = \bar{S}/e\bar{I}$ .

### 3.3 Floquet decomposition

The remaining task is now to compute the stationary solutions  $\rho_0(t)$  and  $\mathcal{F}_{1\perp}(t)$  from the time-dependent equations of motion (54) and (55). Like for the computation of the dc current in our previous work [74], we solve this problem within a Floquet treatment of the isolated wire, which provides a convenient representation of the electron creation and annihilation operators.

#### 3.3.1 Fermionic Floquet operators

In the driven wire Hamiltonian (2), the single-particle contribution commutes with the interaction term and, thus, these two Hamiltonians possess a complete set of common many-particle eigenstates. Here we start by diagonalising the first part of the Hamiltonian which describes the single-particle dynamics determined by the time-periodic matrix elements  $H_{nn'}(t)$ . According to the Floquet theorem, the corresponding (single particle) Schrödinger equation possesses a complete solution of the form

$$|\Psi_\alpha(t)\rangle = e^{-i\epsilon_\alpha t/\hbar} |\varphi_\alpha(t)\rangle, \quad (61)$$

with the so-called quasienergies  $\epsilon_\alpha$  and the  $\mathcal{T}$ -periodic Floquet states

$$|\varphi_\alpha(t)\rangle = \sum_k e^{-ik\Omega t} |\varphi_{\alpha,k}\rangle. \quad (62)$$

The Floquet states and the quasienergies are obtained by solving the eigenvalue problem

$$\left( \sum_{n,n'} n |H_{nn'}(t)\langle n'| - i\hbar \frac{d}{dt} \right) |\varphi_\alpha(t)\rangle = \epsilon_\alpha |\varphi_\alpha(t)\rangle, \quad (63)$$

whose solution allows one to construct via Slater determinants many-particle Floquet states. In analogy to the quasimomenta in Bloch theory for spatially periodic potentials, the quasienergies  $\epsilon_\alpha$  come in classes

$$\epsilon_{\alpha,k} = \epsilon_\alpha + k\hbar\Omega, \quad k \in \mathbb{Z}, \quad (64)$$

of which all members represent the same physical solution of the Schrödinger equation. Thus we can restrict ourselves to states within one Brillouin zone like for example  $0 \leq \epsilon_\alpha < \hbar\Omega$ .

For the numerical computation of the operators  $\rho_0$  and  $\mathcal{F}_{1\perp}$ , it is essential to have an explicit expression for the interaction picture representation of the wire operators. It can be obtained from the (fermionic) Floquet creation and annihilation operators [16] defined via the transformation

$$c_{\alpha s}(t) = \sum_n \langle \varphi_\alpha(t) | n \rangle c_{ns}. \quad (65)$$

The inverse transformation

$$c_{ns} = \sum_\alpha \langle n | \varphi_\alpha(t) \rangle c_{\alpha s}(t) \quad (66)$$

follows from the mutual orthogonality and the completeness of the Floquet states at equal times [70]. Note that the right-hand side of Eq. (66) becomes time independent after the summation. The Floquet annihilation operator (65) has the interaction picture representation

$$\tilde{c}_{\alpha s}(t, t') = U_0^\dagger(t, t') c_{\alpha s}(t) U_0(t, t') \quad (67)$$

$$= e^{-i(\epsilon_\alpha + U N_{\text{wire}})(t-t')/\hbar} c_{\alpha s}(t'), \quad (68)$$

with the important feature that the time difference  $t - t'$  enters only via the exponential prefactor. This allows us to evaluate the  $\tau$ -integration of the master equation (54) after a Floquet decomposition. Relation (68) can easily be shown by computing the time derivative with respect to  $t$  which by use of the Floquet equation (63) becomes

$$\frac{d}{dt} \tilde{c}_{\alpha s}(t, t') = -\frac{i}{\hbar} (\epsilon_\alpha + U N_{\text{wire}}) \tilde{c}_{\alpha s}(t, t'). \quad (69)$$

Together with the initial condition  $\tilde{c}_\alpha(t', t') = c_\alpha(t')$  follows relation (68). Note that the time evolution induced by  $\mathcal{H}_{\text{wire}}(t)$  conserves the number of electrons on the wire.

### 3.3.2 Master equation and current formula

In order to make use of the Floquet ansatz, we decompose the master equation (54) and the current formula (57) into the Floquet basis derived in the last subsection. For that purpose we use the fact that we are finally interested in the current at asymptotically large times in the limit of large interaction  $U$ . The latter has the consequence that only wire states with at most one excess electron play a role, so that the stationary density operator  $\rho_0(t)$  can be decomposed into the  $2N + 1$  dimensional basis  $\{|0\rangle, c_{\alpha s}^\dagger(t) |0\rangle\}$ , where  $|0\rangle$  denotes the wire state in the absence of excess electrons and  $s = \uparrow, \downarrow$ . Moreover, it can be shown that at large times, the density operator becomes diagonal in the electron number  $N_{\text{wire}}$ , so that a proper ansatz reads

$$\rho_0(t) = |0\rangle \rho_{00}(t) \langle 0| + \sum_{\alpha, \beta, s, s'} c_{\alpha s}^\dagger |0\rangle \rho_{\alpha s, \beta s'}(t) \langle 0| c_{\beta s'}. \quad (70)$$

Note that we keep terms with  $\alpha \neq \beta$ , which means that we work beyond a rotating-wave approximation. Indeed in a non-equilibrium situation, the off-diagonal density matrix elements  $\rho_{\alpha\beta}$  will not vanish and neglecting them might lead to artefacts [16, 75].

By inserting the decomposition (70) into the master equation (54), we obtain an equation of motion for the matrix elements  $\rho_{\alpha s, \beta s'} = \langle 0 | c_{\alpha s} \rho_{\text{wire}} c_{\beta s'}^\dagger | 0 \rangle$ . We evaluate the trace over the lead states and compute the matrix element  $\langle 0 | c_{\alpha s}(t) \dots c_{\beta s'}^\dagger(t) | 0 \rangle$ . Thereby we neglect the two-particle terms which are of the structure  $c_{\alpha s}^\dagger c_{\beta s}^\dagger | 0 \rangle \langle 0 | c_{\beta s} c_{\alpha s}$ . Formally, these terms drop out in the limit of strong Coulomb repulsion because they are accompanied by a rapidly oscillating phase factor  $\exp(-iU N_{\text{wire}} \tau / \hbar)$ . Then the  $\tau$ -integration results in a factor  $f_L(\epsilon_{\alpha, k} + U)$  which vanishes in the limit of large  $U$ . Since the total Hamiltonian (1) is diagonal in the spin index  $s$ , we find that the density matrix elements  $\rho_{\alpha s, \beta s'}$  are spin-independent as well so that after a transient stage

$$\rho_{\alpha \uparrow, \beta \uparrow}(t) = \rho_{\alpha \downarrow, \beta \downarrow}(t) \equiv \rho_{\alpha \beta}(t) \quad (71)$$

and  $\rho_{\alpha \uparrow, \beta \downarrow} = 0$ . Moreover, the stationary density operator (70) obeys the time periodicity of the driving field [16] and, thus, can be decomposed into the Fourier series

$$\rho_{\alpha \beta}(t) = \sum_k e^{-ik\Omega t} \rho_{\alpha \beta, k} \quad (72)$$

and  $\rho_{00}(t)$  accordingly.

After some algebra, we arrive at a set of  $N^2$  coupled equations of motion for  $\rho_{\alpha \beta}(t)$  which in Fourier representation read

$$\begin{aligned} i(\epsilon_\alpha - \epsilon_\beta - k\hbar\Omega)\rho_{\alpha \beta, k} = & \frac{\Gamma_L}{2} \sum_{k', k''} \langle \varphi_{\alpha, k' + k''} | 1 \rangle \langle 1 | \varphi_{\beta, k + k''} \rangle \rho_{00, k'} (f_L(\epsilon_{\alpha, k' + k''}) + f_L(\epsilon_{\beta, k + k''})) \\ & - \frac{\Gamma_L}{2} \sum_{\alpha', k', k''} \langle \varphi_{\alpha, k' + k''} | 1 \rangle \langle 1 | \varphi_{\alpha', k + k''} \rangle \rho_{\alpha' \beta, k'} \bar{f}(\epsilon_{\alpha', k + k''}) \\ & - \frac{\Gamma_L}{2} \sum_{\beta', k', k''} \langle \varphi_{\beta', k' + k''} | 1 \rangle \langle 1 | \varphi_{\beta, k + k''} \rangle \rho_{\alpha \beta', k'} \bar{f}(\epsilon_{\beta', k' + k''}) \\ & + \text{same terms with the replacement } 1, L \rightarrow N, R. \end{aligned} \quad (73)$$

In order to solve these equations, we have to eliminate  $\rho_{00, k}$  which is most conveniently done by inserting the Fourier representation of the normalisation condition

$$\text{tr}_{\text{wire}} \rho_0(t) = \rho_{00}(t) + 2 \sum_{\alpha} \rho_{\alpha \alpha}(t) = 1. \quad (74)$$

In order to obtain for the current an expression that is consistent with the restriction to one excess electron, we compute the expectation values in the current formula (57) with the reduced density operator (70) and insert the Floquet representation (66) of the wire operators. Performing an average over the driving period, we obtain for the dc current the expression [74]

$$\begin{aligned} \bar{I} = \frac{2e\Gamma_L}{\hbar} \text{Re} \sum_{\alpha, k} \left( \sum_{\beta, k'} \langle \varphi_{\beta, k' + k} | 1 \rangle \langle 1 | \varphi_{\alpha, k} \rangle \rho_{\alpha \beta, k'} \bar{f}_L(\epsilon_{\alpha, k}) \right. \\ \left. - \sum_{k'} \langle \varphi_{\alpha, k' + k} | 1 \rangle \langle 1 | \varphi_{\alpha, k} \rangle \rho_{00, k'} f_L(\epsilon_{\alpha, k}) \right). \end{aligned} \quad (75)$$

Physically, the second contribution of the current formula (75) describes the tunnelling of an electron from the left lead to the wire and, thus, is proportional to  $\rho_{00} f_L$  which denotes the probability that a lead state is occupied while the wire is empty. The first terms corresponds to the reversed process namely the tunnelling on an electron from site  $|1\rangle$  to the left lead.

The decomposition of the equation of motion (59) for the long-time limit of  $\mathcal{F}_{1\perp}$  and the subsequent computation of the  $\bar{S}$  from Eq. (60) proceeds along the same lines with the only difference that the current operators  $\mathcal{J}_{\pm}$  yield an inhomogeneity and that the r.h.s. of the trace condition (74) is

$$\text{tr}_{\text{wire}} \mathcal{F}_{1\perp} = (\mathcal{F}_{1\perp})_{00} + 2 \sum_{\alpha} (\mathcal{F}_{1\perp})_{\alpha\alpha} = 0. \quad (76)$$

### 3.4 Spinless electrons

A particular consequence of strong Coulomb repulsion is the mutual blocking of different spin channels. This motivates us to also compare to the case of spinless electrons which is physically realised by spin polarisation. For spinless electrons, we drop in the Hamiltonian (1) all spin indices. Physically, this limit is realised by a sufficiently strong magnetic field that polarises all electrons contributing to the transport. By the same calculation as above, we then obtain for the current also the expression (75) but without the prefactor 2. The factor 2 is also no longer present in the normalisation condition (74) which now reads

$$\text{tr}_{\text{wire}} \rho_0(t) = \rho_{00}(t) + \sum_{\alpha} \rho_{\alpha\alpha}(t) = 1, \quad (77)$$

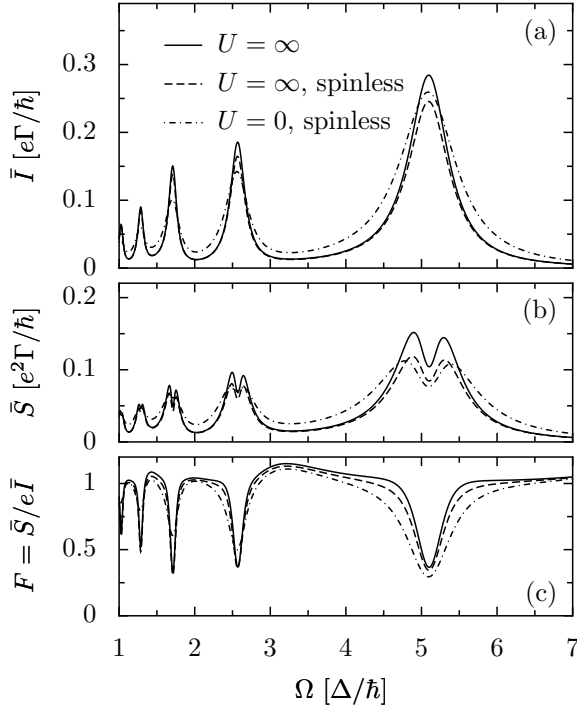
and accordingly in the equation of motion for  $\mathcal{F}_{1\perp}$ .

## 4 Noise in non-adiabatic electron pumps

In a mesoscopic conductor, a spatial asymmetry together with an ac driving can induce a pump current, i.e. a dc current that flows even in the absence of any net bias [21–23]. More precisely, the central necessary condition for this effect is the absence of a particular symmetry, namely generalised parity defined as the invariance under spatial reflection in combination with a time shift by half a driving period [16]. For adiabatically slow driving, the transferred charge is determined by the area enclosed in parameter space during one cycle of the periodic time-evolution [24, 25]. This implies that the resulting dc current, apart from non-adiabatic corrections, is proportional to the driving frequency. For time-reversal symmetric conductors, the parameters do not enclose a finite area and, thus, one finds for small driving frequencies that the pump current obeys  $\bar{I} \propto \Omega^2$  [28].

In both the absence and the presence of time-reversal symmetry, the current increases with the driving frequency, which suggests that pumping is more effective beyond the adiabatic regime. Non-adiabatic electron pumping is particularly interesting because at internal resonances of the central system the pump current can assume rather large values [3, 12, 13]. In the absence of electron-electron interactions, the pump current, in addition, exhibits resonance peaks with a remarkably low current noise [28] and the question arises whether this favourable property persists once Coulomb repulsion becomes relevant. We consider the setup sketched in Fig. 1 and described by the Hamiltonian (1) and compare the results for non-interacting electrons with those for strong Coulomb repulsion. Thereby we focus on the parameter set for which recently a low Fano factor has been predicted [28]: It is characterised by a large internal bias, intermediately strong wire-lead coupling, and resonant driving. Moreover at the prime resonance, the driving amplitude has to fulfil the condition  $J_1(A/\hbar\Omega) = \sqrt{5/3}\Gamma_{\text{L,R}}$ , where  $J_1$  denotes the first-order Bessel function of the first kind.

Figure 2a shows the current obtained for non-interacting electrons and for strong Coulomb repulsion with and without considering the spin degree of freedom. The dc current exhibits characteristic peaks whenever the  $k$ -th-order resonance condition  $\Omega = [(\epsilon_1 - \epsilon_2)^2 + \Delta^2]^{1/2}/k\hbar$  is fulfilled. Interestingly enough, the magnitude of the current is practically the same for all three cases: The interaction yields at most differences of the order 10%. We also find that for  $U = \infty$ , the resonance peaks are slightly sharper, in particular if one takes the electron spin into account. This behaviour has also been found for photo-assisted transport in molecular wires, where it is even significantly more pronounced [74]. The zero-frequency noise shown in Fig. 2b possesses a more involved double-peak structure with a local minimum



**Fig. 2** DC current (a), zero-frequency noise (b), and Fano factor (c) for the non-adiabatically driven electron pump sketched in Fig. 1 as a function of the driving frequency. The dot levels possess the energies  $\epsilon_{1,2} = \pm 2.5\Delta$  and are coupled to the leads with the strength  $\Gamma_{L,R} = 0.3\Delta$ . The driving amplitude is  $A = 3.7\Delta$  and the temperature  $k_B T = 0.005\Delta$ . The peak at  $\Omega \approx \Delta/\hbar$  corresponds to the first-order resonance, while the peaks at lower frequencies are higher-order resonances.

at the centre. This results in a rather pronounced minimum of the Fano factor (Fig. 2c) which assumes values as low as  $F \approx 0.25$ , which means sub-Poissonian current noise.

Our observations lead us to the conclusion that interactions are not an essential obstacle for tuning the double-dot electron pump into a low-noise regime as suggested in Ref. [28]. The reason for this is that for the pump configuration sketched in Fig. 1, one energy level lies below the Fermi energy while the other lies well above. Consequently in equilibrium for a sufficiently small wire-lead coupling, the left site is occupied while the right site is empty, whatever the interaction strength. Thus, the double dot is populated with only one electron so that interactions become irrelevant. Unless the driving amplitude is huge, this occupation is altered only slightly. Consequently interactions do not modify the current significantly. We emphasise that for strong dot-lead coupling  $\Gamma$  and finite interaction  $U$ , these arguments no longer hold true.

In the experiment of Ref. [2], a typical inter-dot coupling is  $\Delta = 50\mu\text{eV}$ . Then, an internal bias  $\epsilon_0 = 5\Delta$  corresponds to the resonance frequency  $\Omega = 5\Delta/\hbar \approx 2\pi \times 60\text{GHz}$ . In both interacting and non-interacting electrons, a wire-lead coupling  $\Gamma = 0.1\Delta$  results in an optimised pump current of the order 200pA with a Fano factor  $F \approx 0.25$ .

## 5 Conclusions

Within the present work, we were particularly interested in the question whether Coulomb repulsion would deteriorate the desirable noise properties of the pump current found for non-interacting electrons. As a central result, we found that very strong interactions do not alter the picture, but merely lead to slight modifications of the current and the zero-frequency noise, which are of the order of 10%. Therefore the essential conclusion of Ref. [28] holds also true in the strongly interacting case: Low-noise operation of a non-adiabatic electron pump requires a large internal bias in combination with a strong inter-dot coupling and resonant driving. The consequence is that a properly tuned driving amplitude provides a relatively large pump current with clearly sub-Poissonian noise. Furthermore, we found that the influence of the electron spin is also not significant.

For non-interacting electrons, the transport problem can be treated within a Floquet scattering formalism, which relates the dc current to the transmission probabilities of elastic and inelastic transmission channels. The latter account for the possibility of photon emission and absorption by a scattered electron. The zero-frequency noise, by contrast, is not only determined by probabilities, but also depends on the scattering phases.

As soon as electron-electron interactions play a significant role, a scattering approach becomes of limited use. In the case of weak wire-lead coupling, the transport problem can then be treated within a master equation approach that is based on second order perturbation theory in the coupling. While previous master equation studies of driven transport are restricted to the computation of the dc current, we developed a method to also compute the noise properties. Our method is based on a time-dependent generalised master equation for the full counting statistics. Concerning its solution, the time-dependence of the Liouville operator represents a particular challenge, because it renders the asymptotic state of the system time-dependent as well. We coped with this difficulty by decomposing the master equation into a proper Floquet basis.

Concerning the noise of non-adiabatic electron pumps, still a couple of intriguing questions remain. Thus far, we revealed that the two limiting cases of vanishing and strong interaction allow for low-noise pumping, but the transition between these limits may nevertheless bear surprises. Thus studying current noise for a finite interaction strength is highly desirable. Moreover, albeit for ac driven conductors experimentally beyond the present state of the art, cumulants of higher order and eventually the full counting statistics can be of interest. In that spirit, our study can only be a first step towards a more complete characterisation of the electron transport in the presence of Coulomb interaction and time-dependent fields.

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